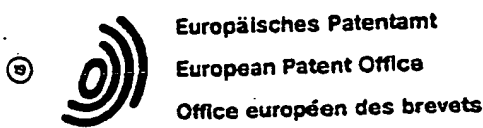


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Unit packaging detergent.

The present invention relates to a unit packaging detergent which is packaged with a water soluble polymer film or sheet and which can be used in water in its packaged form, which is especially improved in respect of the time required for the solubilization of the polymer film. The unit packaging detergent is rapidly dispersed into water and no insoluble polymer film remains on washed clothes.

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EP 0 388 105 A2

UNIT PACKAGING DETERGENT

The present invention relates to a unit packaging detergent and a film suitable for the packaging. The term "unit packaging detergent" in the present specification means a detergent packaged in a water soluble polymer film or sheet in such a form that it is suitable for a one time use.

Hitherto, detergent powders or granules have usually been sold filled in a box or a water-insoluble bag. for instance a polyethylene bag, and a suitable amount of the detergent powder or granules is measured by a spoon or a measuring cup or poured into a washing machine. However, in this conventional manner it is not only inconvenient to use the detergent in a suitable amount for the washing so as not to waste it, but also liable to spill it onto a floor. In addition, since coin operated laundries have recently been developed, the development of a unit packaging detergent to be used therein has been desired.

Hitherto, a unit packaging detergent has been studied and proposed, for instance, the use of polyvinyl alcohol as a film for packaging (Japanese Patent Application KOKAI No. 62-4800), but the solubility of the film in water is insufficient so that a gel or jelly of insoluble polyvinyl alcohol adheres to the washed clothes, or it takes a long time to wash the gel or jelly out of the clothes. Therefore, this method has seldom been practised.

We have now developed a unit packaging detergent which is readily soluble in water and does not remain on the clothes after washing, and a film or a sheet suitable for this purpose.

Accordingly, the present invention provides a unit packaging detergent which comprises a detergent powder or granules packaged with a water soluble film or sheet made of a polymer having the following properties:

- (1) a tear strength of more than about 50 kg/cm,
- (2) a dissolution time of less than about 20 seconds, and
- (3) an equilibrium moisture content of less than about 20% at 20°C and at a relative humidity of 80%.

The present invention also provides a water soluble film or sheet suitable for a packaging substance of a unit packaging detergent, which is made of a polymer having the following properties.

- (1) a tear strength of more than about 50 kg/cm,
- (2) a dissolution time of less than about 20 seconds, and
- (3) an equilibrium moisture content of less than about 20% at 20°C and at a relative humidity of 80%.

The water soluble polymer film or sheet used in the the present invention is prepared from a water soluble polymer having the aforementioned properties.

The tear strength is determined according to JIS (Japanese Industrial Standard) K 6301-1975.

The dissolution time of the polymer in the present specification is defined as the time required for a polymer film having a thickness of 30 μ m and an area of 1 cm² to dissolve completely in water when it is placed on the surface of water at 20°C.

The equilibrium moisture content of the polymer is determined as follows: (1) a test film is placed in a Petri dish, and the Petri dish is put in a desiccator in the bottom of which a saturated aqueous solution of ammonium chloride is contained in an amount of about 5 - 10% by volume of the bottom space, whereby the relative humidity in the desiccator is maintained at about 80% at 20°C; (2) the desiccator is held in a thermostatic chamber at 20°C for 10 days; and (3) the film in the desiccator is weighed daily over these 10 days to determine the equilibrium moisture content of the polymer.

Examples of polymers satisfying the above properties and having film forming properties are polyoxyethylene derivatives, such as polyesters of polyether polyols which contain polyoxyethylene groups, with polycarboxylic acid and the like.

Preferred polyether polyols contain polyoxyethylene groups as an essential element, which may contain other polyoxyalkylene groups derived from alkylene oxides such as propylene oxide, butylene oxide, styrene oxide and the like. Such polyether polyols may be obtained by reacting alkylene oxides with a compound having a plurality of active hydrogen atoms in a molecule (referred to as an active hydrogen-containing compound hereinafter). Such an active hydrogen-containing compound may include alkylene glycols such as ethylene glycol, propylene glycol, butylene glycol, hexylene glycol and the like; aliphatic polyols such as glycerin, sorbitan, pentaerythritol, sugar alcohol, polyglycerin, and the like; aromatic polyols such as hydroquinone, phloroglucinol and the like; amines such as ethylamine, hexylenediamine, phenylenediamine, cyclohexylamine, benzylamine, aniline, imidazolidine and the like; amides; mercaptans; and so on. When the active hydrogen-containing compound contains hetero atoms such as amines, amides or mercaptans, the compound so-obtained is also included in the term "a polyether polyol" as used in this

specification even though it contains hetero atoms.

A preferred polyether polyol is a polyether diol, from the viewpoint of solubility in water and polymeric ability. A monoalcohol cannot extend the polymer chain, and a triol or a polyol having more than 3 hydroxyl groups tends to be reticulated and to be insoluble in water.

Mixtures of the polyether polyols, for example, polyethylene glycol and polypropylene glycol; polyethylene glycol and polyoxyethylene/polyoxypropylene copolymer; polyoxyethylene glycerin and polyoxyethylene/polyoxypropylene copolymer; polyoxyethylene triethanolamine and polyoxyethylene/polyoxypropylene copolymer may also be used. These combinations are not restricted to the above.

The polyethylene polyol itself may contain additional groups such as oxyethylene groups or oxypropylene groups, such may be block copolymers or random copolymers.

The polyether polyol may be obtained according to conventional methods, for example, by reacting a desired alkylene oxide, or a mixture of two or more thereof, with one or more active hydrogen containing compound(s) in the presence of catalyst, for example, an alkaline catalyst such as potassium hydroxide or sodium carbonate, or acid catalyst such as boron trifluoride, under a suitable pressure such as 0 to 10 atoms (gauge pressure) at a temperature of, for example, from 60 to 160° C.

An alkaline catalyst for an addition polymerization of the alkylene oxide is particularly preferred, because the alkaline catalyst can be also be used as the catalyst for a successive esterification without any elimination thereof or use of additional catalyst.

The content of the polyoxyethylene moiety in the polyether polyol is preferably more than 50% by weight, more preferably 70 to 100% by weight based on the total weight of the polyether polyol. If the content of the polyoxyethylene moiety is less than 50%, the water-solubility becomes insufficient.

The polyether polyol should be water soluble, and preferably has a weight average molecular weight of more than about 1,000, more preferably more than about 2,000, more preferably 300 to 50,000 from the viewpoint of its water solubility and tear strength.

As aforementioned, the most preferred examples of the polymers for the present invention are polyesters of polyether polyols with polycarboxylic acids. Such polymers can be prepared according to Japanese Patent Application KOKAI No. 56-226018.

Examples of the polycarboxylic acids which may be used for the preparation of the polyester are aliphatic polycarboxylic acids such as malonic acid, succinic acid, glutaric acid, adipic acid, pimelic acid, suberic acid, azelaic acid, sebacic acid, maleic acid, fumaric acid, mesaconic acid, malic acid, tartaric acid, hexane tricarboxylic acid, and the like; aromatic polycarboxylic acids such as phthalic acid, isophthalic acid, terephthalic acid, trimellitic acid, pyromellitic acid, 1,5-naphthalenedicarboxylic acid, dimmer acid and the like. The polycarboxylic acid may be a partial or complete ester, which can be reacted by an ester interchange reaction, or may be an acid anhydride such as maleic anhydride, phthalic anhydride, pyromellitic dianhydride, and the like. Lower alkyl esters of the polycarboxylic acids are also usable, such as the methyl esters, polymethyl esters, mono or polyethyl esters, and the like.

Mixture of polycarboxylic acids may be used.

Most preferred polycarboxylic acids are phthalic acid, terephthalic acid, isophthalic acid, sebacic acid, pyromellitic acid, lower alkyl esters thereof such as dimethyl esters, and the like.

A preferred weight average molecular weight of the polymer such as polyesters of the polyether polyols and the polycarboxylic acid is preferably in the range of from 10,000 to 500,000. If the molecular weight is less than 10,000, the water-solubility of the polymer film or sheet so obtained would be sufficient, but the tear strength will be lower than the limit required and the equilibrium moisture content will exceed the upper limit required, whereas with a molecular weight of more than 500,000 the water-solubility will decrease.

The ratio of the polyether polyol to the polycarboxylic acid, ester or acid anhydride is not restrictive providing the molecular weight of the polyether polyol is not less than 1,000, and that of the obtained polymer is not less than 10,000 and not more than 500,000.

A film or a sheet obtained from the above polymers, has an excellent water-solubility, and dissolves rapidly in water, that is, it can be dissolved at a speed of about 2 to 5 times that of a water soluble polyvinyl alcohol film of the same thickness. Accordingly, a unit packaging detergent packaged in the film or sheet made of the above polymers is rapidly and completely dissolved in water, when thrown into a washing machine, and causes no problems such as an insoluble polymer adhering to or depositing on the washed clothes. Further, the film or sheet made from the above polymer has such a high tear strength that it is not torn or broken when used as a packaging film or sheet in an autopackaging machine. Further, the film or sheet obtained from the above polymer has such a low equilibrium moisture content that it can maintain a very high strength under a high humidity, as in the rainy season, in spite of its high water-solubility, which is a specifically desirable property. As the aforementioned polymer is chemically stable, and heat sealable,

and additionally the water-solubility of the heat sealed part is maintained so that it does not adhere to and remain on washed cloths after washing, in contrast to PVA, it can be used for unit packaging of a detergent for domestic use or for a vending machine.

The esterification of the polyether polyol and the polycarboxylic acid, ester, or acid anhydride can be effected in a conventional manner, for example, by heating under reduced pressure in the presence of catalyst such as p-toluene sulfonic acid, boron trifluoride, potassium hydroxide, sodium hydroxide, magnesium hydroxide, alkali methals and the like.

The preferred temperature is 80 to 250 °C, and the preferred pressure is 0.001 to 20 Torr. The reaction can be usually completed within 0.5 to 10 hours. Of course, the reaction may be carried out with the removal of generated water or lower alcohol by ventilation with nitrogen gas. The aforementioned esterification is only by way of suitable example, and should not be construed restrictively.

The molecular weight of the polymer may be controlled by the ratio of the polyether polyol to the polycarboxylic acid, or by controlling the reduced pressure. The latter is more practical and easier. As the reaction pressure is lowered, a polymer having a larger molecular weight can be obtained.

Though the aforementioned polyesters are the most suitable polymers for providing a film or a sheet for packaging a detergent, another water soluble polymer such as methyl cellulose, hydroxyethyl cellulose, hydroxypropyl methyl cellulose, carboxy methyl cellulose, polyvinyl alcohol, polyvinylpyrrolidone, polyacrylate (e.g. sodium, potassium or ammonium salts) and the like may be mixed therewith. However, as the polymer of the present invention is immiscible with polyvinyl alcohol, and the co-use with cellulose derivatives makes the film highly water-absorbable, the content of another water soluble polymer is preferably less than 10% by weight of the polymer of the present invention.

A film or a sheet of the polymer suitable for packaging a detergent can be obtained in a conventional manner such as by solvent casting, melt extrusion, and the like.

The film or sheet of the present invention may contain other substances which are usually formulated into detergent compositions, for example, antioxidants, fluorescent agents, defoamers, chelating agents, and the like, or others such as solubility controlling agents, such as a comparatively lower molecular weight polyethylene glycol; plasticizers; mildew-proofing agents; and the like, if desired.

The thickness of the film or sheet should be controlled in respect of the strength or durability to the packaging process, potential use in a vending machine, storage, commercial transportation and the like; water-solubility in use in order to make the detergent dissolve rapidly and to prevent an insoluble substance from redepositing on washed clothes; moisture permeability in order to prevent the detergent from caking, wetting or deteriorating; and the economics thereof.

A usual thickness of the film for the packaging of a detergent is from 10 to 100 µm, more preferably 20 to 60 µm. It would also be preferable to use a film or a sheet having such a thickness in the present invention, because a film or a sheet within this range of thickness is applicable to packaging machines generally used.

The polymers to be used for the film or sheet of the present invention should have (1) a tear strength, (2) a dissolution time and (3) an equilibrium moisture content as aforementioned.

The detergent to be packaged in the water soluble polymer film or sheet of the present invention is not restricted, and may be any usual detergent. The detergent may typically contain surface active agents such as alkylbenzene sulfonate, alkyl sulfonate, soap, polyoxyalkylene alkyl ether, polyoxyalkylene alkylphenyl ether, fatty acid alkylamide and the like; builders, such as polyphosphates, zeolite, carbonates, silicate, organic builders including chelating agents and the like; fluorescent whitening agents; redeposition preventing agents such as sodium silicate, carboxymethylcellulose, hydroxypropyl methyl cellulose, hydroxybutyl methyl cellulose and the like; enzymes such as protease, lipase, amylase and the like; softening agents such as dimethyldialkyl quaternary ammonium salts, imidazolinium compounds and the like; bleaching agents such as sodium percarbonate, sodium perborate and the like; perfumes; bulking agents such as sodium sulfate; defoamers or foam controlling agents; thickening agents; and the like.

The weight of one package (unit) of the unit packaging detergent of the present invention may preferably contain from 3 g to 100 g, more preferably 5 g to 50 g of a detergent. The term "unit packaging" means a pack in which the detergent is packaged in such an amount that it is used in a standard one time wash, but it does not mean the exclusion of two or more packs of detergent used in a one time wash, because the amount of the detergent to be used depends on the dirtiness of washing, amount of washing, volume of washing machine, hardness of water, temperature of water, kind of detergent and the like. A detergent package which is intentionally prepared so that two or more units are used in a one time wash is also included in the term "unit packaging".

Illustrating the invention are the following examples, which, however, are not be construed as limiting the invention to their details. All parts and percentages in the examples, as well as throughout this

specification are by weight unless otherwise specified.

EXAMPLE 1

(1) Preparation of Polymer:

Ethylene oxide 4,500 parts were introduced dropwise into an deaerated autoclave (5 liter) containing propylene glycol 15 parts over about 11 hs. in the presence of potassium hydroxide 2.2 parts as a catalyst as maintaining the reaction temperature at about 90 - 130 °C and the pressure at about 1 to 2 atoms (gauge pressure) to give a polyether polyol having a weight average molecular weight of 15,000, into which then diethyl isophthalate 68 parts were added and heated at about 180 to 190 °C for 1 h. remaining the same catalyst as removing produced ethyl alcohol under a reduced pressure of about 0.1 Torr to give a polyester having an average molecular weight of about 150,000.

(2) Preparation of Film:

The obtained polyester was dissolved in water to give a 20 % aqueous solution (viscosity: about 10,000 cps at 25 °C), the aqueous solution was cast by a coater, and dried at 80 °C for 0.5 hs. to give an even film of 30 µm after cooling.

(3) Determination of Physical Properties:

Physical properties of the obtained film are determined by the following methods:

A tear strength is determined according to JIS (Japanese Industrial Standard) K 6301-1975.

A dissolution time of the polymer in the present specification is defined as the time required for a polymer film having an area of 1 cm² to completely dissolve in water when it is placed on the surface of water at 20 °C.

An equilibrium moisture content of the polymer is determined by (1) a test film is placed in a Petri dish, and the Petri dish is put in a desiccator in the bottom of which a saturated aqueous solution of ammonium chloride is contained in an amount of about 5 - 10 % by volume of the bottom space, whereby the relative humidity in the desiccator is maintained at about 80% at 20 °C for 10 days, and is held in a thermostatic chamber at 20 °C for 10 days, and the film in the desiccator is weighed daily over these 10 days to determine the equilibrium moisture content of the polymer.

The results are:

tear strength: 105 kg/cm,

dissolution time (20 °C): 8 seconds, and

equilibrium moisture content (at 20 °C and at a relative humidity of 80 %) : 10 %

(4) Preparation of Unit Packaging Detergent:

A powder detergent containing sodium salt of a linear alkylbenzene sulfonic acid as a main surfactant (New Beads: trade name of Kao K.K.) was packaged in the film obtained in the process (2) as containing the detergent of 20 g in one package to yield a unit packaging detergent.

(5) Evaluation of Unit Packaging Detergent

Thrown into water (20 °C), the unit packaging detergent obtained in the process (4) was completely dissolved within 10 seconds and the detergent was immediately dispersed into water.

The unit packaging detergent was packed in a cardboard case as occupying about 80 % volume (remaining about 20 % space), and subjected to a practical test (packing, carrying, storing and the like in a commercial route). Neither broken nor wetting package was observed.

EXAMPLE 2(1) Preparation of Polyester:

5 Ethylene oxide 4,000 parts were introduced into a deaerated autoclave (5 liter) containing polyethylene glycol (MW = 3,000) 400 parts in the presence of sodium hydride 2.4 parts over 20 hs. at about 115 °C under a pressure of 1.1 atoms. (gauge pressure) to proceed the addition polymerization of the ethylene oxide to the polyethylene glycol to give a polyethylene glycol having a weight molecular weight of about
 10 32,000, into which dimethyl terephthalate 32 parts was added and reacted at about 200 °C for 1 hs. as removing methanol generated under a reduced pressure of 0.05 Torr. to yield a polyester having an weight average molecular weight of about 250,000.

(2) Preparation of Film:

15 Film of 50 µm thick was prepared from the polyester obtained in the process (1) of the Example 2 using a calender roll.

The film has following properties:

20 tear strength: 150 kg/cm,
 dissolution time (20 °C): 15 seconds at 50 µm thick, 8 seconds at 30 µm thick and
 equilibrium moisture content (at 20 °C and at a relative humidity of 80 %) : 7 %

(3) Preparation of Unit Packaging Detergent and Evaluation:

25 According the same manner as in the Example 1 except that the polyester film of the Example 2 was used instead of that of the Example 1 a unit packaging detergent was prepared and evaluated. The same results as in the Example 1 were obtained.

COMPARATIVE EXAMPLE 1

35 The powder detergent (New Beads) was packaged using a film of 30 µm thick made of polyvinyl alcohol soluble in cold water according to the same manner as in the Example 1. The film has following physical properties:

tear strength: 180 kg/cm, and
 equilibrium moisture content (at 20 °C and at a relative humidity of 80 %) : 13 %
 but the dissolution time (at 20 °C) is 35 seconds.

40 According the same manner as in the Example 1 except that the polyvinyl alcohol film of the above was used instead of polyester film of the Example 1 a unit packaging detergent was prepared and evaluated.

Though the body of the film was completely dissolved in water within comparatively short time, the welded or lapped parts were coagulated in the water to form a gel-like mass, and it adhered on washed clothes.

EXAMPLES 3 - 5

50 According to a similar manner to the Example 1 three kinds of water soluble film were obtained, and the physical properties were determined. The kinds of polyether polyol and polycarboxylic acid used, and the properties are shown in Table 1.

Table 1

EX	polyether polyol		polycarboxylic acid	polymer and property			
	MW.	EO content		MW	(1)	(2)	(3)
1	3000	100 wt %	pyromellitic dianhydride	30000	85	4	17
2	5000	90 wt %	dimethyl sebacate	50000	125	6	12
3.	40000	100 wt %	dimethyl phthalate	300000	170	15	8
EO: polyoxyethylene residue							

(1) a tear strength

(2) dissolution time of the polymer (sec/30 μ m thick)

(3) an equilibrium moisture content

Claims

1. A unit packaging detergent which comprises a detergent powder or granules packaged with a water soluble film or sheet made of a polymer having the following properties:

(1) a tear strength of more than about 50 kg/cm,

(2) a dissolution time of less than about 20 seconds, and

(3) an equilibrium moisture content of less than about 20% at 20° C and at a relative humidity of 80%.

2. A unit packaging detergent as claimed in claim 1 in which the water soluble film has a thickness in the range of from 10 to 100 micrometres.

3. A unit packaging detergent as claimed in claim 1 or claim 2 in which the polymer comprises a polyester of a polyether polyol.

4. A unit packaging detergent as claimed in claim 3 in which the polyester has a weight average weight molecular in the range of from 10,000 to 500,000.

5. A unit packaging detergent as claimed in claim 3 in which the polyester contains polyether polyol groups of a weight average molecular weight of more than about 1,000.

6. A unit packaging detergent as claimed in any one of claims 3 to 5 in which the polymer additionally comprises a water soluble polymer other than the polyester.

7. A water soluble film or sheet suitable for a packaging substance of a unit packaging detergent, which is made of a polymer having the following properties.

(1) a tear strength of more than about 50 kg/cm,

(2) a dissolution time of less than about 20 seconds, and

(3) an equilibrium moisture content of less than about 20% at 20° C and at a relative humidity of 80%.

8. A film or sheet as claimed in claim 7 which has a thickness of in the range of from 10 to 100 micrometres.

9. A film or sheet as claimed in claim 7 in which the polymer comprises a polyester of a polyether polyol.

10. A film or sheet as claimed in claim 9, in which the polyester has a weight average weight molecular in the range of from 10,000 to 500,000.

11. A film or sheet as claimed in claim 9 or claim 10 in which the polyester contains polyether polyol groups of a weight average molecular weight of more than about 1,000.

12. A film or sheet as claimed in any one of claims 9 to 11 in which the polymer additionally comprises a water soluble polymer other than the polyester.



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54 Unit packaging detergent.

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EUROPEAN SEARCH REPORT

Application Number

EP 90 30 2581

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claims	CLASSIFICATION OF THE APPLICATION (Int. Cl.8)
Y	US-A-3 892 905 (R.E. ALBERT) * Whole document *	1,2,7,8	C 11 D 17/04
Y	US-A-3 277 009 (M. FREIFELD et al.) * Whole document *	1,2,7,8	
Y	US-A-3 413 229 (Th.S. BLANCO et al.) * Column 3, lines 15-30; examples *	1,2,7,8	
A	CA-A-1 112 534 (PROCTER & GAMBLE) * Claims 1-4 *	1	
			TECHNICAL FIELDS SEARCHED (Int. Cl.8)
			C 11 D
The present search report has been drawn up for all claims			
Place of search		Date of completion of search	Examiner
The Hague		30 August 91	GOLLER P.
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